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Collisional loss rate in a magneto-optical trap for sodium atoms: Light-intensity dependence

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We have measured the total collisional loss rate for ultracold sodium atoms held in a magneto-optical trap (MOT) as a function of light intensity in the trap. We extract the rate constant for collisional loss by measuring the temporal behavior of MOT loading from background vapor. The loss rate increases with light intensity in satisfactory agreement with new calculations, which are also presented. The results are interpreted in terms of detailed collision processes.

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When the duration of a two-body collision becomes long compared to the time for spontaneous radiative decay, the system must be considered "open" and dissipatively coupled to the bath of empty modes of the radiation field. We report here studies of collisions between sodium atoms in a magneto-optical trap (MOT) that belong to this novel physical domain. In earlier work we considered photoassociative ionization [1], but here we turn our attention to exoergic processes leading to escape from the trap. These processes limit the density and confinement time of three-dimensional optical traps and the ultimate "brightness" of two-dimensionally confined atomic beams. The nature of these ultracold collisional interactions is therefore crucial to the design of dense sources of cold atoms. Here we determine absolute rate constants for collisional trap loss as a function of light intensity in the MOT, and interpret the results in terms of the principal two-body collision processes responsible. Further, we present new calculations from recently developed theory [2,3] of ultracold processes, which we compare to the experimental results.

Earlier investigations [4,5] appreciated the significance of collisional loss to ultimate trap density, and explored the manipulation of trap-loss rate with optical "catalysis." Recent experimental studies [6,7] have refined and extended the early work, confirming the important (but not as yet clearly delineated) role played by hyperfine structure. At the same time, theory has revealed the extreme sensitivity of any quantal calculation to the scattering potential data of the problem [8]. Meaningful comparison of experiment and theory must be restricted to those systems for which the relevant scattering potentials are known with spectroscopic precision. Therefore we have concentrated our efforts on sodium collisions where the ground and first singly excited states are known with sufficient accuracy.

We report here a study of collision loss in a magneto-optical trap for sodium atoms, showing the rate constant dependence on light intensity in a range from 10 to 150 $\mathrm{mW\,cm^{-2}}$. Our measurement technique is similar to the one of Ref. [6], which exploits the radiation trapping properties of an optically thick cloud of trapped atoms. The MOT operates in a closed vapor cell, and the trap is loaded with atoms from the low-velocity tail present in the vapor. Three mutually orthogonal retroreflected laser beams, tuned $\simeq 10$ MHz to the red of an atomic transition frequency, intersect at the center of a quadrupolar magnetic field generated by a pair of current-carrying coils. At the trap center, the magnetic field is zero and grows linearly in all directions. The Zeeman splitting of the atomic levels and the use of appropriate laser polarizations produce a spatially dependent light force with a net effect of restoring the atomic position to trap center where the magnetic field is null. In addition to the trapping force, the red detuning produces a viscous, damping force, cooling the trapped atoms to about 300 μK. The cell is a stainless-steel chamber containing a partial pressure of sodium vapor at 353 K, with a background base pressure maintained below 10^{-6} Pa (10^{-8} Torr). The magnetic-field coils are located external to the chamber and produce an axial field gradient of about 0.002 $\mathrm{T\,cm^{-1}}$ (20 $\mathrm{G\,cm^{-1}}$).

Light from a monomode-ring dye laser passes through an electro-optic modulator, introducing red and blue sidebands that are separated from the carrier by 1712 MHz, before dividing into the three beams of equal intensity along the orthogonal trapping axes.

As in Ref. [4] we observe trapping for two different tunings of the laser. A type-I trap results from tuning the carrier laser frequency close to the $3s\ ^2S_{1/2}(F=2)\rightarrow 3p\ ^2P_{3/2}(F'=3)$ transition and the blue sideband near the $3s\ ^2S_{1/2}(F=1)\rightarrow 3p\ ^2P_{3/2}(F'=2)$ transition. The carrier drives the principal MOT trapping and cooling transition, while the blue sideband functions as a "repumper" to prevent population loss from the main optical cycle. A type-II trap is ob-

tained with the carrier frequency tuned close to $3s\,^2S_{1/2}(F=2)\rightarrow 3p\,^2P_{3/2}(F'=2)$ and the blue sideband to $3s\,^2S_{1/2}(F=1)\rightarrow 3p\,^2P_{3/2}(F'=0)$. Figure 1 illustrates both tuning conditions. The type-I configuration produces a robust trap with a typical volume of 10^{-4} cm^3 and density $\sim 10^{10}$ atoms cm^{-3}. In contrast, type II is weaker [9], with a typical volume of $\sim 5\times 10^{-3}$ cm^3 and density $\sim 10^{8}$ atoms cm^{-3}. We normally work with a type-I trap, and the loss rates reported here refer to this arrangement. We determine the trap density by imaging the fluorescent volume onto a calibrated photomultiplier tube (PMT) while measuring its dimensions ($\simeq 200\times 600$ μm^2) with a telescope mounted on precision XY translation stages. Knowledge of the fluorescence rate f striking the PMT and the solid angle subtended determines the atom total number N through the relation

$$f=GN\frac{\rho_{22}}{\tau}=GN\frac{1}{2\tau}\frac{\Omega_0^2/2}{(\omega_0-\omega)^2+(\Gamma/2)^2+(\Omega_0^2/2)}\ , \qquad (1)$$

where G is the photon collection efficiency; N the total number of atoms in the trap; ρ_{22} the fractional excited-state population; and τ, Γ, and Ω_0 the radiative lifetime, natural linewidth, and on-resonant Rabi frequency, respectively.

We write β as the rate constant for trap loss due to ultracold collisions in the trap and α the rate constant for trap loss due to collisions of trapped atoms with "hot" background gas atoms and molecules. The loss rate from ultracold collisions and from background gas collisions is then, respectively, $\beta n^2(r,t)$ and $\alpha n_b n(r,t)$, with $n(r,t)$ the spatially and temporally varying local trap density and n_b the background density. It is convenient to absorb αn_b into a single constant, γ. As observed by Walker, Sesko, and Wieman, [10] in cesium, and confirmed by us for sodium, trap loading proceeds at constant density, i.e., as the number of atoms loading into the trap increases, the volume expands so that the density in the trap remains unchanged. As a result, the equation that governs the total number N of atoms in the trap can be written as

$$dN/dt=L-(\gamma+\beta n_c)N\ , \qquad (2)$$

where L is the loading rate and n_c is the fixed atomic density achieved in the trap under constant conditions. The solution of this equation is an exponential represented by $N=N_0\{1-\exp(-\gamma-\beta n_c)t\}$. A semilog plot of $(1-N/N_0)$ versus t determines the slope equal to $-(\gamma+\beta n_c)$. In Fig. 2 we show typical loading curves (fluorescence versus time) for several laser intensities of the main trapping frequency. From fitting the loading curves we extract the semilog plots as shown in Fig. 3. These plots are indeed linear, which confirms that trap loading takes place within the regime of constant density as described above. In fitting the loading curves we ignore very early times where the constant density regime has not yet been reached. The family of slopes in Fig. 3 shows the light field intensity dependence of $(\gamma+\beta n_c)$. The variation of slope with field intensity must be associated with changes in the term β, since γ and n_c are both constant. We measure n_c in the experiment; in order to determine β, it remains to devise an independent measurement of γ.

For this purpose we have used the type-II trap tuned $\simeq 30$ MHz to the red from the resonance condition. In a type-II trap the atom density is sufficiently small that background collisions dominate trap loss, and the slope of the semilog plot of Fig. 3 is equal only to γ. The slope should be independent of trap density, light intensity, or magnetic field. We have tested the insensitivity of γ to these parameters by carrying out auxiliary experiments. In contrast, by changing the cell temperature we have shown that γ is proportional to the background pressure, as expected. Values of γ have also been determined using two other independent techniques [11], and the three methods result in good agreement.

We have measured β over a range of laser intensities, and the results of several independent runs are shown in Fig. 4. Despite error bars of 50% or more, the dependence of the rate constant on light intensity is unmistakable. The main source of error in β is the trap volume determination. Careful scrutiny of the trapped-atom spatial distribution reveals an approximate oblate spheroid, but the detailed shape varies slightly from run to run, thereby introducing the consequent uncertainty in the

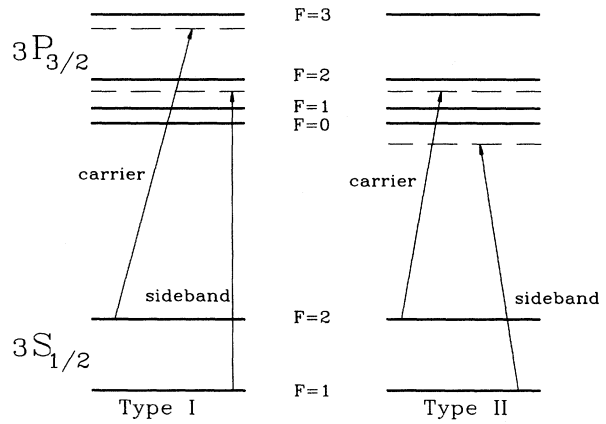

FIG. 1. Tuning schemes for type-I and type-II traps. Dashed lines in type I indicate detuning $\simeq 10$ MHz to the red of resonance. The sideband (repumper) in type II is detuned 30 MHz to the red of resonance. The carrier acts as the trapping and cooling transition and the sideband acts as the repumper in both types of MOT.

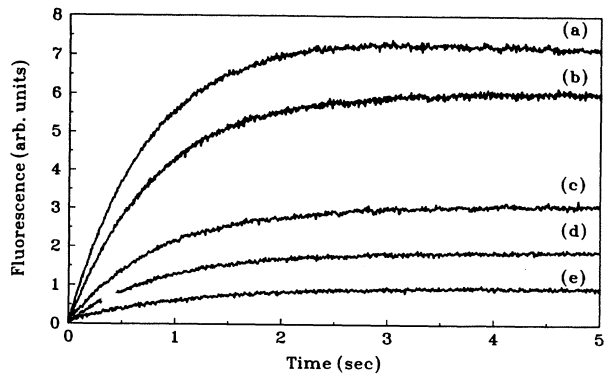

FIG. 2. Loading curves of the trap for several light intensities.

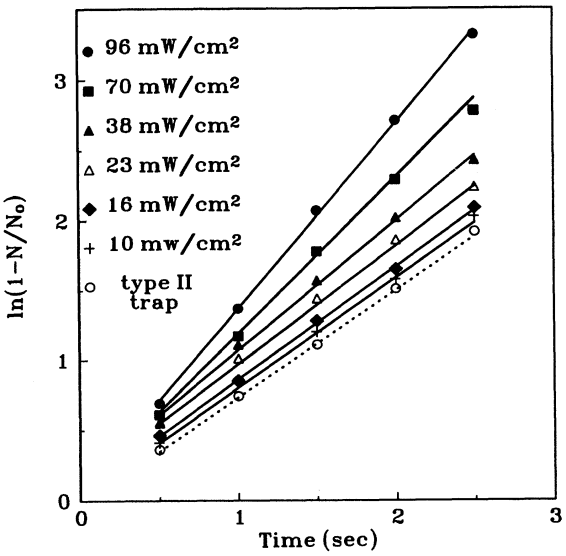

FIG. 3. Logarithmic plot [$|\ln(1-N/N_0)|$ vs time] for the loading curves of Fig. 2. The dashed line represents the plot for a type-II trap.

trap-loss rate constant. The error bars reflect an upper and lower bound on the trap volume. The light intensity dependence of the rate constant only really becomes evident over a much wider range than that investigated in Ref. [4] ($\simeq 10-50$ mW cm^{-2}). Systematic measurement errors masking the relatively modest variation in β in this narrow range may have contributed to the erroneous conclusion that the loss mechanism is independent of the excited-state atomic fraction. The measured value for the rate constant in Ref. [4] ($\beta=4\times10^{-11}$ cm^3 sec^{-1}), while greater than the present results by about a factor of 2 (within the same intensity range), agrees within their quoted range of uncertainty of a factor of 5.

Julienne and Vigué [2] (JV) extended the local equilibrium theory of Gallagher and Pritchard [12] to calculate the detailed molecular mechanisms responsible for collisional Na trap loss due to radiative escape (RE) and fine-structure changing (FS) collisions. The theory neglects the hyperfine structure of the Na_2 molecule, a possibly serious approximation, which we address below. Both processes arise from the collision between a ground-state and an excited-state atom. The RE process is essentially a free-free molecular transition in which some fraction of final ground-state atom pairs possesses enough kinetic energy to escape the trap. The FS collision results in a change of fine-structure level. In the case of sodium, collisions between a ground-state atom and an excited atom in the $3p\ ^2P_{3/2}$ level result in a transition to the $3p\ ^2P_{1/2}$ level, with the concomitant exoergic release of 17 cm^{-1} of kinetic energy. Every FS or RE collision results in the loss of two atoms from the trap. The JV[2] expression for the rate of trap loss through RE or FS collisions is

$$K_{\text{loss}}=\tfrac{1}{4}(v\pi/k^2)\sum_l(2l+1)P_xP_{ES}\ , \qquad (3)$$

where v is the velocity, k the momentum wave vector, the

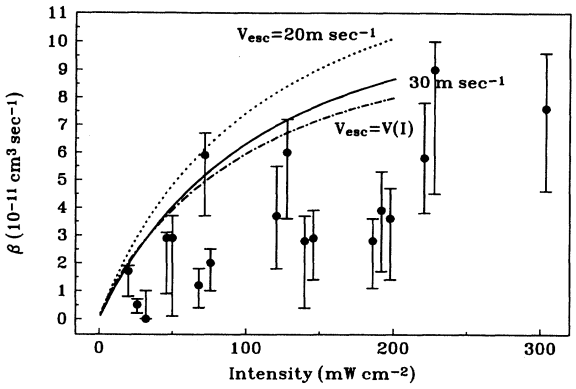

FIG. 4. Trap-loss rate coefficient as a function of intensity for the type-I trap. Different symbols represent different runs of our experiment. The full lines represent theory calculation using the model described in this paper.

factor of $\frac{1}{4}$ is due to ground-state degeneracy, neglecting hyperfine structure, l is the relative angular momentum quantum number, P_x the probability in a short-range molecular basis set of the x=FS or RE process for a given molecular state, and P_{ES} the probability of exciting the upper molecular state at long range and surviving relative to spontaneous emission as the atoms come together and reach a distance short enough to undergo the x process. Instead of the local equilibrium model of JV, we have used the new optical Bloch equation (OBE) method of Band and Julienne [3] to calculate P_{ES} as the population density matrix element for the upper molecular state. The probabilities P_x for the RE process were calculated for the attractive 0_u^+, 0_g^-, and 1_g states using the formula in Eq. (50) of Ref. [12], and the contribution summed according to Eq. (3). One difficulty in comparing experiment with theory is to know the escape velocity criterion to apply for RE; i.e., how much minimum velocity must be imparted to the ground-state atoms in order for them to escape the confining forces of the MOT? Assuming single-atom escape velocities of 20 and 30 m/s, RE is calculated to contribute 38% and 24%, respectively, of the total loss rate at low power. We have also constructed a model in which the escape velocity is calculated versus MOT laser intensity, varying between about 20 and 40 m/s over the experimental intensity range. This range is consistent with previous MOT studies [13–15].

We have reevaluated the fine-structure changing probability P_x of Ref. [12] using new quantum close-coupling calculations, performed within a total angular momentum representation in which the electronic-rotational Hamiltonian is asymptotically diagonal. Molecular hyperfine structure due to the nuclear spin is neglected in these calculations. For a given total angular momentum J, and for each gerade (g) and ungerade (u) manifold of Na_2, there are six coupled channels for each molecular parity. In the present work, we have used the new accurate potential curves of Magnier *et al.* [16] for the four molecular states [in Hund's case (a)] correlated with the $3s+3p$ dissociation limit, instead of the model curves used previously [2]. Only the u manifold contributes significantly to the FS process, through two attractive entrance channels, 0_u^+ and 2_u, that correlate to the $^2P_{3/2}$

state. The calculations include the spin-orbit interaction between $^1\Sigma_u^+$ and $^3\Pi_u$ states at their short-range crossing at 8 a.u. and the Coriolis mixing of the three components $\Omega=0,1,2$ of the $^3\Pi_u$ state. The main contribution for Na comes from Coriolis coupling. At 300 μK the contribution from the 2_u entrance channel is dominant, because the scattering flux on this state has an excellent survival probability due to its very long lifetime. Radiative decay of the 2_u state is dipole forbidden and only occurs at large internuclear separation by virtue of retardation corrections. Excitation of the 2_u state from the ground state is sufficiently allowed to permit excitation of the atoms when they are far apart, and the small spontaneous-emission rate results in excellent survival of the excited state as the atoms come together. We have scaled the spin-orbit matrix elements in our calculation to the measured $^3\Pi_u$ spin-orbit splitting. Transition probabilities for Na are not very sensitive to this spin-orbit scaling: using the value scaled from the asymptotic atomic splitting only leads to about a 15% increase in the FS rate coefficient. We find similar FS probabilities P_x from the 0_u^+ and 2_u entrance channels, which can be represented according to Ref. [2] as $0.000\,13l(l+1)$. The fact that these probabilities are a factor of 2 smaller than those estimated in Ref. [2] underscores the necessity of utilizing accurate molecular data in scattering calculations.

The agreement between theory and experiment indicated in Fig. 4 is reasonably satisfactory, although theory tends to be on the high side. New experiments [7,17] demonstrate that hyperfine effects are important in Rb MOT's, and there is no reason to expect them to be unimportant for Na as well. Nevertheless, the order-of-magnitude agreement between theory and experiment here, as well as for Cs trap loss [2,3], indicates that some of the qualitative features of the physics is right. Trap loss in Na traps by RE collisions and by FS collisions through the 2_u state will continue to play a role even when hyperfine structure is introduced, though the magnitudes and relative contributions may change. Our calculation of the short-range P_x is independent of asymptotic hyperfine considerations, though the long-range P_{ES} may be quite sensitive to hyperfine-structure effects. Calculations on the role of hyperfine structure are in progress [18]. The OBE method is well suited to the investigation of such effects.

For a detuning of one natural linewidth, which was assumed in Fig. 4 in comparing with experiment, the trap-loss rate coefficient is predominately due to FS processes due to the 2_u entrance channel. But theory predicts a sharp drop in this contribution with increasing red detuning, as the atoms are excited when they are closer together and excitation of the 2_u state becomes more forbidden. Our calculations without hyperfine structure predict that the RE process becomes dominant over the FS process with increasing red detuning, contributing 10%, 38%, 63%, and 78% percent, respectively, to the low-power total trap-loss rate at red detunings of 5, 10, 15, and 20 MHz if a minimum atomic escape velocity of 20 m/s is assumed. This effect could be measured by detecting the rate of production of $^2P_{1/2}$ atoms as a function of detuning. The ratio of $^2P_{1/2}$ production to total trap loss should decrease sharply with increasing red detuning. This effect will be investigated in future experiments.

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[1] V. Bagnato, L. Marcassa, Y. Wang, J. Weiner, P. S. Julienne, and Y. B. Band (unpublished).

[2] P. S. Julienne and J. Vigué, Phys. Rev. A **44**, 4464 (1991).

[3] Y. B. Band and P. S. Julienne, Phys. Rev. A **46**, 330 (1992).

[4] M. Prentiss *et al.*, Opt. Lett. **13**, 452 (1988).

[5] D. Sesko *et al.*, Phys. Rev. Lett. **63**, 961 (1989).

[6] D. Hoffmann *et al.*, Phys. Rev. Lett. **69**, 753 (1992).

[7] C. D. Wallace *et al.*, Phys. Rev. Lett. **69**, 897 (1992).

[8] O. Dulieu, P. Julienne, and J. Weiner, in Abstracts of the Workshop on Ultracold Collision Dynamics, Institute of Theoretical Atomic, and Molecular Physics, Cambridge, Massachusetts (1992) (available from Institute of Theoretical Atomic and Molecular Physics).

[9] A complete investigation of the type-II trap has recently been carried out by D. Milori *et al.* (unpublished).

[10] T. Walker *et al.*, Phys. Rev. Lett. **64**, 408 (1990).

[11] Using a type-I trap at very low light intensity, far-red detuning, and low magnetic field, γ can be measured by measuring trap loading at very early times. In addition, γ can be determined by extrapolating the slope of Fig. 3 as laser intensity goes to zero. Further details of these techniques will be presented in a later publication.

[12] A. Gallagher and D. E. Pritchard, Phys. Rev. Lett. **63**, 957 (1989).

[13] E. L. Raab *et al.*, Phys. Rev. Lett. **59**, 2631 (1987).

[14] C. Monroe *et al.*, Phys. Rev. Lett. **65**, 1571 (1990).

[15] K. E. Gibble *et al.*, Opt. Lett. **17**, 526 (1992).

[16] S. Magnier, Ph. Millie, O. Dulieu, and F. Masnou-Seeuws, J. Chem. Phys. (to be published).

[17] T. Walker (unpublished).

[18] C. J. Williams, P. S. Julienne, and Y. B. Band (unpublished).